

This Page Is Inserted by IFW Operations  
and is not a part of the Official Record

## **BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning documents *will not* correct images,  
please do not report the images to the  
Image Problem Mailbox.**

# PATENT SPECIFICATION

(11) 1 504 172

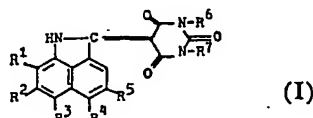
1 504 172

- (21) Application No. 20270/75 (22) Filed 14 May 1975  
 (31) Convention Application No. 2 428 198  
 (32) Filed 11 June 1974 in  
 (33) Fed. Rep. of Germany (DE)  
 (44) Complete Specification published 15 March 1978  
 (51) INT CL<sup>2</sup> C09B 57/00, 5/30, 49/12  
 (52) Index at acceptance  
 C4P 104 I22 D1G1 D1G D1M3 D1M Q2L1 Q2X

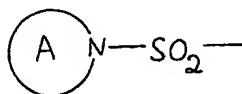


## (54) NAPHTHOLACTAM DYES

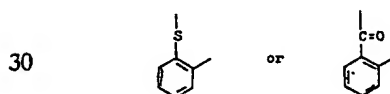
- (71) We, BASF AKTIENGESSELLSCHAFT, a German Joint Stock Company, of 6700 Ludwigshafen, Federal Republic of Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—  
 The invention relates to dyes of the formula  
 (I):



- in which  
 R<sup>1</sup> is hydrogen, chloro, bromo, alkyl, alkoxy, nitro or arylmercapto;  
 R<sup>2</sup> is hydrogen or chloro;  
 R<sup>3</sup> is hydrogen, chloro, bromo, alkyl, alkoxy, nitro, carboxylic acylamino, alkylsulfonylamino, arylsulfonylamino, alkylmercapto, arylmercapto, arylsulfonyl, alkylsulfonyl, unsubstituted or substituted sulfamoyl, alkanoyl, aroyl or a heterocyclic group of the formula:—



- in which the ring A is a saturated heterocyclic group;  
 R<sup>4</sup> is hydrogen, chloro, alkoxy or arylmercapto; or  
 R<sup>3</sup> and R<sup>4</sup> together form a radical of the formula:



- R<sup>5</sup> is hydrogen, chloro or alkoxy; and  
 R<sup>6</sup> and R<sup>7</sup> independently of one another are

alkyl; alkyl bearing hydroxy, cyano, alkoxy or carbalkoxy as a substituent; phenyl; or phenyl bearing chloro, methyl or methoxy as a substituent, at least one of R<sup>1</sup> to R<sup>5</sup> being different from hydrogen.

Interpretations of R<sup>1</sup> to R<sup>5</sup>:

It is to be understood that references herein to aryl moieties (e.g. in arylmercapto or in aroyl) are intended to cover substituted aryl moieties as well as unsubstituted aryl moieties.

Examples of alkyl, alkoxy and arylmercapto groups for R<sup>1</sup> are methyl, ethyl, methoxy, ethoxy, phenylmercapto or phenylmercapto bearing chloro, methyl, methoxy, phenyl, phenoxy or methoxycarbonyl as a substituent.

Examples for R<sup>3</sup> are the same radicals as for R<sup>1</sup> and also naphthylmercapto, acetylaminophenylmercapto, acetyl-amino, propionyl-amino, benzoylamino, benzoylamino bearing chloro, methyl or methoxy as a substituent, methylsulfonylamino, ethylsulfonylamino, phenylsulfonylamino, tolylsulfonylamino, methylmercapto,  $\beta$  - hydroxyethylmercapto, methylsulfonyl, ethylsulfonyl, phenylsulfonyl, tolylsulfonyl, chlorophenylsulfonyl, acetyl, propionyl, butyryl, benzoyl, benzoyl bearing methyl, methoxy, chloro or bromo as a substituent, sulfamoyl, N - methylsulfamoyl, N - ethylsulfamoyl, N - butylsulfamoyl, N - phenylsulfamoyl, N - chlorophenylsulfamoyl, N - methylphenylsulfamoyl, N - methoxyphenylsulfamoyl, N - trifluoromethylphenylsulfamoyl, N - methyl - N - phenylsulfamoyl, N,N - dimethylsulfamoyl, N,N - diethylsulfamoyl, N,N - dipropylsulfamoyl, N,N - dibutylsulfamoyl, pyrrolidinosulfonyl, piperidinosulfonyl or morpholinosulfonyl. Radicals of the formulae: NHCONH<sub>2</sub>, NHCONHCH<sub>3</sub>, and NHCONHC<sub>6</sub>H<sub>5</sub> are also suitable.

Examples of radicals R<sup>4</sup> are hydrogen, chloro and the alkoxy and arylmercapto radicals specified for R<sup>1</sup>.

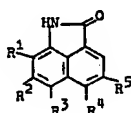
Examples of alkoxy radicals R<sup>5</sup> are methoxy and ethoxy.

Particular examples of R<sup>6</sup> and R<sup>7</sup> are alkyl of one to four carbon atoms, alkoxyalkyl of three to eight carbon atoms, hydroxyalkyl of two or three carbon atoms, cyanoethyl, alkoxy-

carbonylalkyl of one to four carbon atoms in the alkoxy and also phenyl.

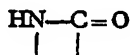
Specific examples are: propyl, butyl, methoxyethyl, ethoxyethyl, butoxyethyl, methoxypropyl, ethoxypropyl, pentoxypropyl,  $\beta$ -hydroxyethyl,  $\beta$ -hydroxypropyl, methoxycarbonylethyl, ethoxycarbonylethyl and butoxycarbonylethyl and preferably methyl, ethyl and phenyl.

Dyes of the formula (I) may be prepared by reacting a naphtholactam of the formula (II):

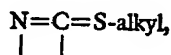
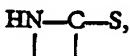


(II)

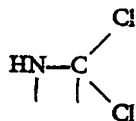
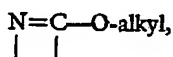
or a derivative of the same in which the grouping



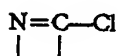
is replaced by



20



or



with an  $N,N'$ -disubstituted barbituric acid derivative of the formula (III)



(III)

Compounds of the formula (II) are known from the literature or may be prepared by methods analogous to those described in the literature.

The reaction of the naphtholactams with the compounds of formula (III) proceeds in the presence of a condensing agent; when the said naphtholactam derivatives are used the presence of a condensing agent may be dispensed with.

Phosphorus halides such as phosphorus

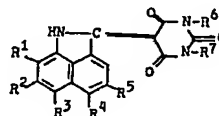
pentachloride, phosphorus trichloride or phosphorus oxytribromide and particularly phosphorus oxytrichloride are suitable as condensing agents.

The reaction may be carried out in an inert solvent such as toluene, chlorobenzene, dichlorobenzene, nitrobenzene or dioxane or also in an excess of the condensing agent.

When the reaction is carried out with a naphtholactam derivative and without a condensing agent the abovementioned solvents are again suitable; examples of additional solvents are pyridine, glacial acetic acid, dimethylformamide and  $N$ -methylpyrrolidone.

The reactions are known in principle and details may be taken from the Examples in which parts and percentages are by weight.

The invention relates particularly to dyes of the formula:



in which

$R^1$  is hydrogen, chloro, bromo,  $C_1$  to  $C_4$  alkyl, methoxy, ethoxy, nitro, phenylmercapto or phenylmercapto bearing chloro, methyl or methoxy as a substituent;

$R^2$  is hydrogen or chloro;

$R^3$  is chloro, bromo,  $C_1$  to  $C_4$  alkyl, methoxy, ethoxy, nitro, acetylamino, propionylamino, benzoylamino, methylsulfonylamino, ethylsulfonylamino, phenylsulfonylamino, tolylsulfonylamino, methylmercapto, ethylmercapto, phenylmercapto, phenylmercapto bearing chloro, methyl or methoxy as a substituent, naphthylmercapto, phenylsulfonyl, phenylsulfonyl bearing chloro or methyl as a substituent, methylsulfonyl, ethylsulfonyl,  $N$ -mono- $C_1$ -to- $C_4$ -alkyl-substituted sulfamoyl,  $N,N$ -di- $C_1$ -to- $C_4$ -alkyl-substituted sulfamoyl,  $N$ -phenylsulfamoyl,  $N$ -chlorophenylsulfamoyl,  $N$ -methylphenylsulfamoyl,  $N$ -methoxyphenylsulfamoyl,  $N$ -trifluoromethylphenylsulfamoyl, pyrrolidonosulfonyl, piperidinosulfonyl, morpholinosulfonyl,  $C_2$  to  $C_4$  alkanoyl, benzoyl, or benzoyl bearing chloro, bromo, methyl, ethyl, methoxy or ethoxy as a substituent;

$R^4$  is hydrogen, chloro, methoxy, ethoxy, phenylmercapto, or phenylmercapto bearing chloro, methyl or methoxy as a substituent; or

$R^5$  and  $R^4$  together are

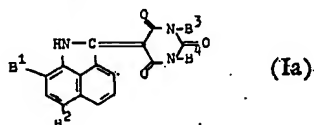


or



$R^6$  is hydrogen, chloro, methoxy or ethoxy; and

$R^6$  and  $R^7$  are independently  $C_1$  to  $C_4$  alkyl,  $C_2$  to  $C_6$  alkyl bearing hydroxy, cyano,  $C_1$  to  $C_4$  alkoxy or  $C_1$  to  $C_4$  alkoxy carbonyl as a substituent, phenyl, or phenyl bearing chloro, methyl or methoxy as a substituent. Particular industrial importance attaches to dyes of formula (Ia):



in which

$B^1$  is hydrogen, chloro, bromo or arylmercapto;

$B^2$  is chloro, bromo, arylmercapto, arylsulfonyl, or aroyl; and

$B^3$  and  $B^4$  are independently methyl, ethyl or phenyl, methyl being particularly preferred.

The following are preferred arylmercapto, arylsulfonyl and aroyl radicals: phenylmercapto, phenylmercapto bearing chloro, methyl or methoxy as a substituent, phenylsulfonyl or benzoyl, or benzoyl or phenylsulfonyl bearing chloro or methyl as a substituent.

Dyes of formula (I) are yellow to violet and have high brilliance and color strength. They are suitable for dyeing synthetic fibers, particularly polyester fibers, and dyeings having very good fastness properties and particularly excellent fastness to light are obtained. Thermal resistance properties may be influenced by the choice of substituents. Dyes which sublime easily are very suitable for transfer printing.

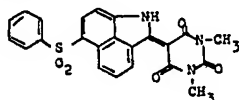
The new dyes are moreover eminently suitable for the mass coloration of plastics; fluorescent colorations having high fastness to light are obtained as a rule.

The invention includes within its scope dye formulations for dyeing synthetic fibers, the formulations containing a dye in accordance with the invention.

The following Examples illustrate the invention.

#### Example 1.

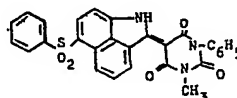
200 parts of phosphorus oxychloride is dripped into a mixture of 309 parts of 4-phenylsulfonylnaphtholactam - (1,8), 165 parts of  $N,N'$ -dimethylbarbituric acid and 1200 parts by volume of toluene at  $90^\circ\text{C}$ . The whole is stirred for 6 hours at  $100^\circ\text{C}$  and 1200 parts by volume of methanol is added during cooling so that the dye is deposited in the form of brown crystals. After cooling, the dye is filtered off, washed with methanol and dried. 357 parts of the dye of the constitution



is obtained which dyes polyester from an aqueous liquor brilliant yellow hues of high tinctorial strength and very good light fastness and thermal stability.

#### Example 2.

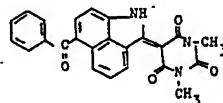
The procedure of Example 1 is followed, but the  $N,N'$ -dimethylbarbituric acid is replaced by an equivalent amount of  $N$ -methyl -  $N$ -phenylbarbituric acid. 427 parts of the dye of the constitution



is obtained which dyes polyester fast golden yellow hues.

#### Example 3.

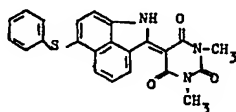
273 parts of 4-benzoylnaphtholactam - (1,8) and 190 parts of  $N,N'$ -dimethylbarbituric acid are introduced into 1000 parts by volume of toluene and stirred at  $95^\circ\text{C}$ . 300 parts of phosphorus oxychloride is dripped in within thirty minutes so that the starting materials pass into solution. The whole is stirred for four hours at  $100^\circ\text{C}$  and during cooling 2000 parts by volume of methanol is allowed to flow in. After suction filtration, washing with methanol and drying there is obtained 360 parts of the dye having the constitution:



in the form of yellowish brown crystals. The dye dyes polyester brilliant greenish yellow shades from an aqueous liquor; the dyeings have very good fastness to dry-heat pleating and setting and to light. In thermoplastics such as polystyrene fluorescent yellow hues having outstanding stability at high temperatures and outstanding fastness to light are obtained.

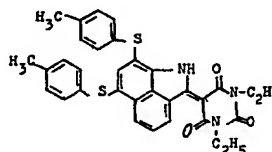
#### Example 4.

250 parts of phosphorus oxychloride is added at  $100^\circ\text{C}$  within one hour to a mixture of 277 parts of 4-phenylmercaptanaphtholactam - (1,8), 190 parts of  $N,N'$ -dimethylbarbituric acid and 900 parts by volume of chlorobenzene. The whole is stirred for another four hours at  $100^\circ\text{C}$  and during cooling there is added a solution of 150 parts of triethylamine in 1500 parts by volume of ethanol. After suction filtration, washing with ethanol and drying there is obtained 349 parts of the dye of the constitution:



in the form of a dark red crystalline powder. Brilliant scarlet dyeings and prints of a high level of fastness properties are obtained on polyesters. Thermoplastics such as polystyrene for example are colored fluorescent scarlet shades having excellent stability at high temperatures and excellent fastness to light.

suction filtered, washed with methanol and dried. 49.2 parts of the dye of the constitution:



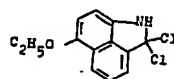
is obtained which gives in polystyrene bluish red hues of outstanding stability at high temperatures and outstanding fastness to light.

#### Example 5.

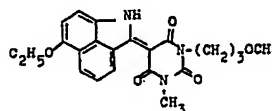
293 parts of 4 - phenylmercaptolactam - (1,8) and 160 parts of N,N' - dimethylbarbituric acid are boiled in 400 parts by volume of N-methylpyrrolidone for eight hours. 200 parts by volume of ethanol is added during cooling. The product is suction filtered, washed and dried. 302 parts of dye is obtained which is identical with that specified in Example 4.

#### Example 8.

268 parts of the compound of the formula



(prepared by the method of Gerinan Laid-Open Specification 1,445,624, Example 2) is heated for 30 minutes at 100 to 120°C in 1,000 parts of xylene. 220 parts of N - methyl - N' - (3 - methoxypropyl) - barbituric acid is added and heating is continued at 100°C for 120 minutes. During cooling, 500 parts of methanol is added and the dye isolated in the usual manner. 311 parts of the dye of the constitution



is obtained in the form of orange crystals, which dye polyester reddish yellow hues having very good lightfastness.

The following dyes of the general formula (I) are prepared according to the methods specified in Examples 1 to 8.

#### Example 6.

307 parts of 4 - phenylmercapto - thionaphtholactam - (1,8) - S - methyl ether and 160 parts of N,N'-dimethylbarbituric acid are boiled in 600 parts by volume of acetic acid for 1 hour. The dye precipitates in the form of red crystals. Upon working up, 328 parts of dye is obtained which is identical with that specified in Example 4.

#### Example 7.

413 parts of 2,4 - bis - (4 - methylphenylmercapto) - naphtholactam - (1,8) and 20 parts of N,N'-diethylbarbituric acid are introduced into 150 parts by volume of dichlorobenzene and stirred at 95°C. 30 parts of phosphorus oxychloride is dripped in so that the components pass into solution. The whole is stirred for six hours at 95°C, diluted with 300 parts by volume of methanol and allowed to cool while stirring. The red crystals are

Ex.	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	Hue
9	-C <sub>2</sub> H <sub>5</sub>	H	H	H	-CH <sub>3</sub>	-CH <sub>3</sub>		greenish yellow
10	H	"	-CH <sub>3</sub>	"	"	"	"	"
11	"	"	Cl	"	"	"	"	reddish yellow
12	"	"	"	"	"	-C <sub>6</sub> H <sub>5</sub>	"	"
13	"	"	"	"	"	-C <sub>6</sub> H <sub>5</sub>	"	"
14	Cl	"	"	"	"	-CH <sub>3</sub>	-CH <sub>3</sub>	"
15	"	"	"	"	"	"	-C <sub>2</sub> H <sub>5</sub>	"
16	Cl	H	Cl	H	-CH <sub>3</sub>	-(CH <sub>2</sub> ) <sub>3</sub> OCH <sub>3</sub>	"	"
17	H	H	H	Cl	-CH <sub>3</sub>	-CH <sub>3</sub>	"	"
18	"	"	"	Cl	"	"	"	orange yellow
19	"	"	"	"	"	-C <sub>2</sub> H <sub>5</sub>	-C <sub>2</sub> H <sub>5</sub>	"
20	"	Cl	"	"	Cl	-CH <sub>3</sub>	-CH <sub>3</sub>	yellowish orange
21	"	"	"	"	"	-C <sub>2</sub> H <sub>5</sub>	-C <sub>2</sub> H <sub>5</sub>	"
22	"	"	"	"	"	-CH <sub>3</sub>	-(CH <sub>2</sub> ) <sub>3</sub> OCH <sub>3</sub>	"
23	"	"	"	"	"	-C <sub>2</sub> H <sub>5</sub>	-C <sub>2</sub> H <sub>5</sub>	"
24	"	H	Br	H	-CH <sub>3</sub>	-CH <sub>3</sub>	-CH <sub>3</sub>	reddish yellow
25	"	"	"	"	"	"	-C <sub>2</sub> H <sub>5</sub>	"
26	"	"	"	"	"	"	-(CH <sub>2</sub> ) <sub>3</sub> OCH <sub>3</sub>	"
27	"	"	"	"	"	-C <sub>6</sub> H <sub>5</sub>	-C <sub>6</sub> H <sub>5</sub>	"
28	Br	"	Cl	"	"	-CH <sub>3</sub>	-CH <sub>3</sub>	"

TABLE (Continued)

Ex.	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	Hue
29	Cl	H	Br	H	H	-CH <sub>3</sub>	-CH <sub>3</sub>	reddish yellow
30	Br	"	"	"	"	"	"	yellowish orange
31	"	"	"	"	"	"	-(CH <sub>2</sub> ) <sub>3</sub> OCH <sub>3</sub>	"
32	"	"	"	"	"	"	-(CH <sub>2</sub> ) <sub>3</sub> OC <sub>2</sub> H <sub>5</sub>	"
33	"	"	"	"	"	-C <sub>4</sub> H <sub>9</sub>	-C <sub>4</sub> H <sub>9</sub>	"
34	"	"	"	"	"	-C <sub>6</sub> H <sub>5</sub>	-C <sub>6</sub> H <sub>5</sub>	"
35	H	H	NO <sub>2</sub>	"	"	-CH <sub>3</sub>	-CH <sub>3</sub>	golden yellow
36	"	"	"	"	"	-C <sub>2</sub> H <sub>5</sub>	-C <sub>2</sub> H <sub>5</sub>	"
37	NO <sub>2</sub>	"	Br	"	"	-CH <sub>3</sub>	-CH <sub>3</sub>	reddish yellow
38	Br	"	NO <sub>2</sub>	"	"	"	"	golden yellow
39	H	H	OCH <sub>3</sub>	"	"	"	"	yellow
40	"	"	"	"	"	-CH <sub>3</sub>	-C <sub>2</sub> H <sub>5</sub>	"
41	"	"	"	"	"	-C <sub>6</sub> H <sub>5</sub>	-C <sub>6</sub> H <sub>5</sub>	"
42	"	"	OC <sub>2</sub> H <sub>5</sub>	"	"	-CH <sub>3</sub>	-CH <sub>3</sub>	"
43	"	"	NHCONH <sub>2</sub>	"	"	"	"	"
44	"	"	"	"	"	-C <sub>6</sub> H <sub>5</sub>	-C <sub>6</sub> H <sub>5</sub>	"
45	"	"	NHCONHCH <sub>3</sub>	"	"	"	"	"
46	"	"	NHCONHC <sub>2</sub> H <sub>5</sub>	"	"	"	"	"
47	"	"	NHCOC <sub>2</sub> H <sub>5</sub>	"	"	-CH <sub>3</sub>	-CH <sub>3</sub>	"

TABLE (Continued)

Ex.	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	Hue
48	H	H	NHCOC <sub>6</sub> H <sub>5</sub>	H	H	-C <sub>2</sub> H <sub>5</sub>	-C <sub>2</sub> H <sub>5</sub>	yellow
49	"	"	NHSO <sub>2</sub> C <sub>6</sub> H <sub>5</sub>	"	"	-CH <sub>3</sub>	-CH <sub>3</sub>	"
50	"	"	NHSO <sub>2</sub> CH <sub>3</sub>	"	"	"	"	"
51	"	"	H	-OCH <sub>3</sub>	"	"	"	"
52	"	"	"	H	-OCH <sub>3</sub>	"	"	"
53	"	"	"	"	-OC <sub>2</sub> H <sub>5</sub>	"	"	"
54	"	"	SCH <sub>3</sub>	"	H	"	"	orange
55	"	"	SC <sub>6</sub> H <sub>5</sub>	"	"	"	-C <sub>2</sub> H <sub>5</sub>	yellowish red
56	"	"	SC <sub>6</sub> H <sub>5</sub>	"	"	"	-(CH <sub>2</sub> ) <sub>3</sub> OCH <sub>3</sub>	"
57	"	"	"	"	"	-C <sub>2</sub> H <sub>5</sub>	-C <sub>2</sub> H <sub>5</sub>	"
58	"	"	"	"	"	-CH <sub>3</sub>	-C <sub>4</sub> H <sub>9</sub>	"
59	"	"	"	"	"	-C <sub>6</sub> H <sub>5</sub>	-C <sub>6</sub> H <sub>5</sub>	"
60	"	"	Cl	SC <sub>6</sub> H <sub>5</sub>	"	-CH <sub>3</sub>	-CH <sub>3</sub>	orange yellow
61	"	"	"	"	"	"	-(CH <sub>2</sub> ) <sub>3</sub> OCH <sub>3</sub>	"
62	"	"	SC <sub>6</sub> H <sub>4</sub> CH <sub>3</sub> (*)	H	"	-CH <sub>3</sub>	-CH <sub>3</sub>	red
63	"	"	"	"	"	-C <sub>2</sub> H <sub>5</sub>	-C <sub>2</sub> H <sub>5</sub>	"
64	"	"	Cl	SC <sub>6</sub> H <sub>4</sub> CH <sub>3</sub> (*)	"	-CH <sub>3</sub>	-CH <sub>3</sub>	orange
65	"	"	SC <sub>6</sub> H <sub>4</sub> OCH <sub>3</sub> (*)	H	"	"	"	red
66	"	"	SC <sub>6</sub> H <sub>4</sub> OCH <sub>3</sub> (*)	"	"	"	-C <sub>4</sub> H <sub>9</sub>	"

TABLE (Continued)

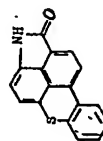
Ex.	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	Hue
67	H	H	SC <sub>6</sub> H <sub>4</sub> OCH <sub>3</sub> (*)	H	H	-CH <sub>3</sub>	-(CH <sub>2</sub> ) <sub>3</sub> OC <sub>2</sub> H <sub>5</sub>	red
68	"	"	Cl	SC <sub>6</sub> H <sub>4</sub> OCH <sub>3</sub> (*)	"	"	-CH <sub>3</sub>	yellowish red
69	"	"	SC <sub>6</sub> H <sub>4</sub> Cl (*)	H	"	"	"	orange
70	"	"	"	"	"	"	-C <sub>4</sub> H <sub>9</sub>	"
71	"	"	SC <sub>6</sub> H <sub>3</sub> Cl <sub>2</sub> (2,5)	"	"	"	-CH <sub>3</sub>	yellowish orange
72	"	"	SC <sub>6</sub> H <sub>4</sub> C <sub>6</sub> H <sub>5</sub> (*)	"	"	"	"	red
73	"	"	SC <sub>6</sub> H <sub>4</sub> OC <sub>6</sub> H <sub>5</sub> (*)	"	"	"	"	"
74	"	"	SC <sub>6</sub> H <sub>4</sub> NHCOCH <sub>3</sub> (*)	"	"	"	"	yellowish red
75	"	"	SC <sub>6</sub> H <sub>4</sub> COOCH <sub>3</sub> (*)	"	"	"	"	red
76	"	"	Naphthyl-2-S-	"	"	"	"	"
77	"	"	"	"	"	-C <sub>2</sub> H <sub>5</sub>	-C <sub>2</sub> H <sub>5</sub>	"
78	SC <sub>6</sub> H <sub>5</sub>	"	Cl	"	"	-CH <sub>3</sub>	-CH <sub>3</sub>	orange
79	"	"	"	"	"	"	-C <sub>4</sub> H <sub>9</sub>	"
80	"	"	NO <sub>2</sub>	"	"	"	-CH <sub>3</sub>	"
81	"	"	SC <sub>6</sub> H <sub>5</sub>	"	"	"	"	reddish orange
82	"	"	"	"	"	-C <sub>4</sub> H <sub>9</sub>	-C <sub>4</sub> H <sub>9</sub>	"
83	"	"	"	"	"	-CH <sub>3</sub>	-(CH <sub>2</sub> ) <sub>3</sub> OC <sub>2</sub> H <sub>5</sub>	"
84	"	"	"	"	"	-C <sub>6</sub> H <sub>5</sub>	-C <sub>6</sub> H <sub>5</sub>	"
85	H	"	"	SC <sub>6</sub> H <sub>5</sub>	"	-CH <sub>3</sub>	-CH <sub>3</sub>	yellowish red

TABLE (Continued)

Ex.	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	Hue
86	H	H	SC <sub>6</sub> H <sub>5</sub>	SC <sub>6</sub> H <sub>5</sub>	H	-CH <sub>3</sub>	-C <sub>2</sub> H <sub>5</sub>	yellowish red
87	SC <sub>6</sub> H <sub>4</sub> CH <sub>3</sub> (*)	"	SC <sub>6</sub> H <sub>4</sub> CH <sub>3</sub> (*)	H	"	"	-CH <sub>3</sub>	red
88	"	"	"	H	"	-CH <sub>3</sub>	-(CH <sub>2</sub> ) <sub>3</sub> OCH <sub>3</sub>	"
89	"	"	"	H	"	-C <sub>4</sub> H <sub>9</sub>	-C <sub>4</sub> H <sub>9</sub>	"
90	SC <sub>6</sub> H <sub>4</sub> OCH <sub>3</sub> (*)	"	SC <sub>6</sub> H <sub>4</sub> OCH <sub>3</sub> (*)	"	"	-CH <sub>3</sub>	-CH <sub>3</sub>	bluish red
91	"	"	"	"	"	-C <sub>2</sub> H <sub>5</sub>	-C <sub>2</sub> H <sub>5</sub>	"
92	H	"	"	SC <sub>6</sub> H <sub>4</sub> OCH <sub>3</sub> (*)	"	-CH <sub>3</sub>	-CH <sub>3</sub>	yellowish red
93	SC <sub>6</sub> H <sub>4</sub> Cl (*)	"	SC <sub>6</sub> H <sub>4</sub> Cl (*)	H	"	"	"	reddish orange
94	SC <sub>6</sub> H <sub>4</sub> Cl (*)	"	"	"	"	"	-C <sub>4</sub> H <sub>9</sub>	"
95	SC <sub>6</sub> H <sub>3</sub> Cl <sub>2</sub> (2,5)	"	SC <sub>6</sub> H <sub>3</sub> Cl <sub>2</sub> (2,5)	"	"	"	-CH <sub>3</sub>	orange

## Example 96.

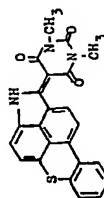
200 parts of N,N'-dimethylbarbituric acid and 275 parts of the compound of the constitution:



5

are brought into solution in 1800 parts by volume of anhydrous nitrobenzene by heating. 220 parts of phosphorus oxychloride is dripped in at 100°C and the whole is stirred for another twelve hours at 100°C. After cooling it is diluted with an equal volume of ethanol, suction filtered, washed with ethanol and dried. 321 parts of the dye of the formula:

10



15




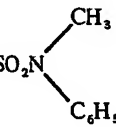
is obtained in the form of a dark crystalline powder. When the dye is incorporated into polystyrene it gives violet hues having good stability to high temperature and good lightfastness properties.

When the N,N'-dimethylbarbituric acid is replaced by barbituric acids having other substituents, dyes having very similar hues are obtained.

Other dyes of the constitution (I) which are obtained analogously to Examples 1 to 8, are:—

20

25

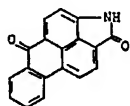
Example	$R^3(R^1=R^2=R^4=R^5=H)$	$R^6$	$R^7$	Hue
97	$-SO_2C_6H_5$	$-CH_3$	$-C_4H_9$	golden yellow
98	"	"	$-(CH_2)_3OC_2H_5$	" "
99	"	$-C_4H_9$	$-C_4H_9$	" "
100	$-SO_2C_6H_4CH_3$ (4)	$-CH_3$	$-CH_3$	" "
101	"	"	$-C_2H_5$	" "
102	"	$-C_6H_5$	$-C_6H_5$	" "
103	$-SO_2C_6H_4Cl$	$-CH_3$	$-CH_3$	" "
104	"	$-C_2H_5$	$-C_2H_5$	" "
105	"	$-CH_3$	$-C_4H_9$	" "
106	"	$-C_4H_9$	$-C_4H_9$	" "
107	$-SO_2N(CH_3)_2$	$-CH_3$	$-CH_3$	yellow
108	$-SO_2N(C_2H_5)_2$	"	"	"
109	$-SO_2N(C_4H_9)_2$	"	"	"
110	$-SO_2N$ 	"	"	"
111	"	"	$-C_2H_5$	"
112	$-SO_2N$ 	"	$-CH_3$	"
113	$-SO_2N$ 	$-C_4H_9$	$-C_4H_9$	"
114	$-SO_2NHC_6H_5$	$-CH_3$	$-CH_3$	"
115	$-SO_2NHC_6H_4CH_3$ (4)	"	"	"
116	$-SO_2NHC_6H_4Cl$ (2)	"	"	"
117	$-SO_2NHC_6H_4OCH_3$ (4)	"	"	"
118	$-SO_2NHC_6H_4CF_3$ (3)	"	"	"
119	$-SO_2N$ 	"	"	"
120	$-COCH_3$	"	"	greenish yellow
121	$-COC_3H_7(iso)$	$-C_6H_5$	$-C_6H_5$	" "
122	$-COC_6H_5$	$-C_2H_5$	$-C_2H_5$	" "
123	"	$-CH_3$	$-C_4H_9$	" "
124	"	"	$-C_6H_5$	" "
125	"	$-C_6H_5$	"	" "

Example	R <sup>3</sup> (R <sup>1</sup> =R <sup>2</sup> =R <sup>4</sup> =R <sup>5</sup> =H)	R <sup>6</sup>	R <sup>7</sup>	Hue
126	-COC <sub>6</sub> H <sub>4</sub> CH <sub>3</sub> (2)	-CH <sub>3</sub>	-CH <sub>3</sub>	yellow
127	-COC <sub>6</sub> H <sub>4</sub> CH <sub>3</sub> (4)	"	"	"
128	"	"	-C <sub>2</sub> H <sub>5</sub>	"
129	-COC <sub>6</sub> H <sub>4</sub> Cl (2)	"	-CH <sub>3</sub>	greenish yellow
130	-COC <sub>6</sub> H <sub>4</sub> Cl (4)	"	"	" "
131	"	-C <sub>2</sub> H <sub>5</sub>	-C <sub>2</sub> H <sub>5</sub>	" "
132	"	-CH <sub>3</sub>	-(CH <sub>2</sub> ) <sub>3</sub> OCH <sub>3</sub>	" "
133	-COC <sub>6</sub> H <sub>3</sub> Cl <sub>2</sub> (2,4)	"	-CH <sub>3</sub>	" "
134	"	"	-C <sub>4</sub> H <sub>9</sub>	" "
135	-COC <sub>6</sub> H <sub>3</sub> Cl <sub>2</sub> (3,4)	"	-CH <sub>3</sub>	" "
136	-COC <sub>6</sub> H <sub>3</sub> Cl <sub>2</sub> (2,5)	"	"	" "
137	"	"	-(CH <sub>2</sub> ) <sub>3</sub> OC <sub>2</sub> H <sub>5</sub>	" "
138	"	-C <sub>6</sub> H <sub>5</sub>	-C <sub>6</sub> H <sub>5</sub>	" "

#### Example 139.

208 parts of N - methyl - N' - butyl-barbituric acid and 271 parts of the compound of the constitution:

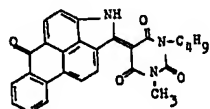
5



are stirred into 1200 parts by volume of anhydrous trichlorobenzene at 100°C. 300 parts of phosphorus oxychloride is dripped in within one hour and the whole is stirred for another eight hours at 100°C to 110°C. During cooling the whole is diluted with 800 parts by volume of methanol and then suction filtered, washed with methanol and dried. 361 parts of the dye of the constitution:

10

15

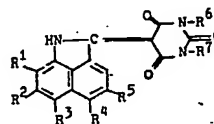


20

is obtained in the form of reddish brown crystals. Luminous orange colorations having good fastness properties are obtained with the dye in thermoplastics, as for example polystyrene.

#### WHAT WE CLAIM IS:—

1. A naphtholactam dye of the formula:



in which

R<sup>1</sup> is hydrogen, chloro, bromo, alkyl, alkoxy, nitro or arylmercapto;

R<sup>2</sup> is hydrogen or chloro;

R<sup>3</sup> is hydrogen, chloro, bromo, alkyl, alkoxy,

nitro, carboxylic acylamino, alkylsulfonyl-

amino, arylsulfonylamino, alkylmercapto,

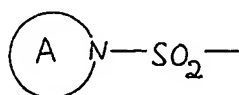
arylmercapto, arylsulfonyl, alkylsulfonyl,

sulfamoyl, sulfamoyl bearing one or two sub-

stituents on the nitrogen atom, alkanoyl,

aroyl or a heterocyclic group of the for-

mula:—



in which the ring A is a saturated heterocyclic group;

R<sup>4</sup> is hydrogen, chloro, alkoxy or arylmercapto; or

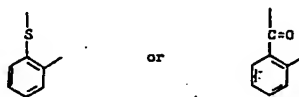
R<sup>3</sup> and R<sup>4</sup> together form a radical of the formula:

25

30

35

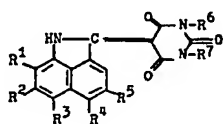
40



$R^5$  is hydrogen, chloro or alkoxy; and  
 $R^6$  and  $R^7$  are independently alkyl, alkyl  
 bearing hydroxy, cyano, alkoxy or carbal-  
 koxy as a substituent, phenyl or phenyl  
 bearing chloro, methyl or methoxy as a  
 substituent;

and at least one of the radicals  $R^1$  to  $R^5$  is  
 other than hydrogen.

2. A dye of the formula:



in which

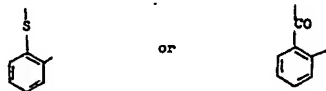
$R^1$  is hydrogen, chloro, bromo,  $C_1$  to  $C_4$  alkyl,  
 methoxy, ethoxy, nitro, phenylmercapto or  
 phenylmercapto bearing chloro, methyl or  
 methoxy as a substituent;

$R^2$  is hydrogen or chloro;

$R^3$  is chloro, bromo,  $C_1$  to  $C_4$  alkyl, methoxy,  
 ethoxy, nitro, acetyl amino, propionyl amino,  
 benzoyl amino, methylsulfonyl amino, ethyl-  
 sulfonyl amino, phenylsulfonyl amino, tolyl-  
 sulfonyl amino, methylmercapto, ethylmer-  
 capto, phenylmercapto, phenylmercapto  
 bearing chloro, methyl or methoxy as a  
 substituent, naphthylmercapto, phenylsul-  
 phonyl, phenylsulfonyl bearing chloro or  
 methyl as a substituent, methylsulfonyl,  
 ethylsulfonyl, N - mono - ( $C_1$  to  $C_8$ ) -  
 alkylsubstituted sulfamoyl, N,N - di - ( $C_1$  to  
 $C_8$ ) - alkyl-substituted sulfamoyl, N -  
 phenylsulfamoyl, N - chlorophenylsulfamoyl,  
 N - methylphenylsulfamoyl, N -  
 methoxyphenylsulfamoyl, N - trifluoro-  
 methylphenylsulfamoyl, pyrrolidinosulfonyl,  
 piperidinosulfonyl, morpholinosulfonyl,  $C_2$   
 to  $C_4$  alkanoyl, benzoyl, or benzoyl bear-  
 ing chloro, bromo, methyl, ethyl, methoxy  
 or ethoxy as a substituent;

$R^4$  is hydrogen, chloro, methoxy, ethoxy,  
 phenylmercapto or phenylmercapto bearing  
 chloro, methyl or methoxy as a substituent;

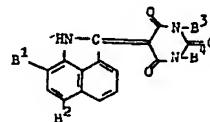
or  
 $R^3$  and  $R^4$  together are:



$R^5$  is hydrogen, chloro, methoxy or ethoxy;  
 and  
 $R^6$  and  $R^7$  are independently  $C_1$  to  $C_4$  alkyl,

$C_2$  to  $C_8$  alkyl bearing hydroxy, cyano,  $C_1$   
 to  $C_4$  alkoxy or  $C_1$  to  $C_4$  alkoxy carbonyl  
 as a substituent, phenyl or phenyl bearing  
 chloro, methyl or methoxy as a substituent.

3. A dye as claimed in claim 1 and having  
 the formula:



in which

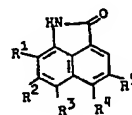
$B^1$  is hydrogen, chloro, bromo or arylmer-  
 capto;

$B^2$  is chloro, bromo, arylmercapto, arylsulfonyl  
 or aroyl; and

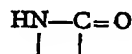
$B^3$  and  $B^4$  independently are methyl, ethyl  
 or phenyl.

4. A dye as claimed in claim 1 and speci-  
 fied in any one of the foregoing Examples.

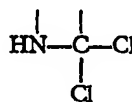
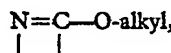
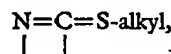
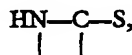
5. A process for the production of a dye  
 as claimed in claim 1 wherein a naphtholactam  
 of the formula:



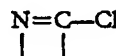
or a derivative of the same in which the group-  
 ing



is replaced by



or



is reacted with an N,N-disubstituted barbituric  
 acid derivative of the formula:



6. A process as claimed in claim 5 carried out substantially as described in any one of the foregoing Examples.

5 7. A dye as claimed in Claim 1 and obtained by a process as claimed in Claim 5 or Claim 6.

8. A dye formulation for dyeing synthetic fibers and which contain a dye as claimed in any of claims 1 to 4 or in Claim 7.

10 9. Plastics material whenever coloured by the presence therein of a dye as claimed in

any of claims 1 to 4 or in Claim 7.

10. Synthetic fibers which have been dyed with a dye formulation as claimed in claim 8.

J. Y. & G. W. JOHNSON,  
Furnival House,  
14—18 High Holborn,  
London WC1V 6DE,  
Chartered Patent Agents,  
Agents for the Applicants.

Printed for Her Majesty's Stationery Office by the Courier Press, Leamington Spa, 1978.  
Published by the Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from  
which copies may be obtained.